TECHNICAL REPORT

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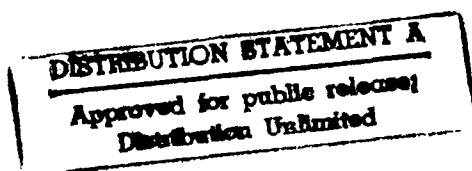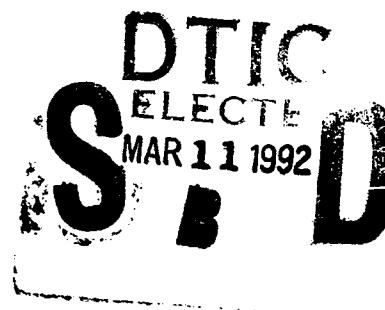
By

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Thermoplastic resin composites undergo a substantial temperature drop during their post-manufacturing cool-down and sustain substantial residual stresses due to mutual geometric constraints among the multi-directional plies. In view of the time-dependent thermomechanical response of the resin the residual stresses exhibit strong dependence on cool-down history. This paper demonstrates that it is possible to obtain an optimal cool-down path which minimizes the residual thermal stresses in APC-2 composites.

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RESIDUAL THERMAL STRESSES IN GRAPHITE/PEEK (APC-2) LAMINATES

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Abstract

Thermoplastic resin composites undergo a substantial temperature drop during their post-manufacturing cool-down and sustain substantial residual stresses due to mutual geometric constraints among the multi-directional plies. In view of the time-dependent thermomechanical response of the resin the residual stresses exhibit strong dependence on cool-down history. This paper demonstrates that it is possible to obtain an optimal cool-down path which minimizes the residual thermal stresses in APC-2 composites.

1. INTRODUCTION

APC-2 (graphite/PEEK) composites exhibit substantial time-dependent stress-strain response, especially at high temperatures, and a very large disparity between the longitudinal and transverse coefficients of thermal expansion. In addition, the processing of APC-2 involves cool-down from $T_M \approx 400°C$ to $T_R \approx 20°C$ - which is about twice the temperature excursion sustained by graphite/ epoxy composites. The above factors produce significant residual thermal stresses in multi-directionally reinforced laminates, which exceed the range of linear behavior. In view of the time-dependent response of PEEK, these stresses are sensitive to temperature history.

It seems that the most complete characterization to date of the time-dependent response of PEEK and unidirectionally reinforced APC-2 coupons was obtained by Xiao [1], and typical creep data at various levels of temperature are shown in Figure 1. These, and supplementary experimental results not shown here, enabled Xiao to represent the time dependent response of PEEK and APC-2 by means of the non-linear viscoelastic model of Schapery [2]. However, it is important to note that the creep data in ref. [1] were collected under isothermal conditions and are limited to the temperature range of $20°C<T<200°C$. It was shown in ref. [3] that the implementation of Schapery's model requires additional creep data under transient temperature conditions. Consequently, Xiao's characterization contains an uncertain component. The time-dependent response of APC-2 laminates can be predicted from the behavior of a single ply by means of classical laminate theory [4] upon the utilization of Schapery's model and employment of the quasi-elastic viscoelastic approximation [5].

When linearly viscoelastic materials undergo a prescribed, geometrically constrained temperature drop over a finite, predetermined, time interval it is possible to find an optimal cool-down path $T_\Omega(t)$ which results in minimal residual stresses. Such paths were found for thermorheologically simple [6] - [8] and thermorheologically complex [9] viscoelastic responses. However, the foregoing analyses do not apply to APC-2 composites because of the substantial non-linearity which occurs during their cool-down. Consequently, the extension of the optimization scheme to account for non-linear behavior is the subject of the present work.

2. ANALYSIS

2.1 Basic Equations

The linear thermo-elastic response of a uni-directionally reinforced ply undergoing a temperature excursion $\Delta T = T - T_1$ from some stress-free reference temperature T_1 (say), is given by [4]

2

$$\varepsilon_L - \alpha_L \Delta_T = \sigma_L/E_L - \nu_{TL}\,\sigma_T/E_T \quad , \quad \varepsilon_T - \alpha_T\Delta T = -\nu_{LT}\,\sigma_L/E_L + \sigma_T/E_T \tag{1}$$

where standard symbols were used and subscripts L and T denote longitudinal and transverse directions, respectively. In the particular case of a symmetric balanced cross-ply lay-up a straight forward employment of laminate theory yields the following expression for the laminate-level residual thermal stresses $\sigma_L = -\sigma_T = -\sigma$:

$$\sigma = - r\, E_T\, \alpha\Delta T \tag{2}$$

where

$$\frac{1}{r} = (1 + \nu_{TL})\left(1 + \frac{1 + \nu_{LT}}{1 + \nu_{TL}}\frac{E_T}{E_L}\right) \quad \text{and} \quad \alpha = \alpha_T - \alpha_L \tag{3}$$

Note that the form of equation (2) remains valid for other symmetric lay-ups (in particular, quasi-isotropic lay-ups), but with different expressions for r. In the case of APC-2, the most pronounced time dependence occurs in the transverse modulus E_T. However, since $E_T/E_L \ll 1$, we may ignore the time dependence of r and let r = 0.9125 to within ± 2%. In the isothermal case with the stress free temperature denoted by T_I, where T = constant and $\Delta T = T_I - T$, employment of Schapery's non-linear viscoelastic model modifies equation (2) to read as follows [2]:

$$\sigma(t) = - r\, \alpha V_2(T)\int_0^t E\big(\xi(t) - \xi(\tau)\big)\left[d\big(V_1(T)\Delta T\big)/d\tau\right]d\tau \tag{4}$$

where the product $V(T) = V_1(T)V_2(T)$ is the vertical shift required to coalesce isothermal data (say creep) to form a "master curve."* In addition, the "reduced times", $\xi(t)$ and $\xi(\tau)$ in equation (4) are given by

$$\xi(u) = \int_0^u dp/a(T; \sigma(p)) \tag{5}$$

In equation (5) $a(T;\sigma)$ is the horizontal shift factor required to coalesce isothermal data (say creep) to form a "master curve." Though both "V" and "a" may, in principle, depend on σ - it appears that the non-linearity of the response can be expressed through $a(T;\sigma)$ alone. Under fluctuating temperatures, with T = T(t), it is necessary to re-write equation (4) with $V_2(T(t))$ and $V_1(T(\tau))$. In this case we also have $\Delta T = \Delta T(\tau)$ in equation (4) and $a = a(T(p);\sigma(p))$ in equation (5).

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* It is obvious from equation (4) that for T = constant σ(t) depends only on V(T) and not on $V_1(T)$ and $V_2(T)$ separately. This no longer holds for fluctuating temperatures.

2.2 Reduction of APC-2 Response Data

The isothermal creep data shown in Figure 1 were reduced by Xiao [1] to expressions which fit Schapery's model. However, the present analysis requires values which involve the relaxation modulus E(t), rather than creep compliance D(t). E(t) was obtained from creep-response functions by the well known relation [5]:

$$E(t) = \frac{1}{D(t)} \frac{\sin(\pi p(t))}{\pi p(t)} \qquad \text{where} \qquad p(t) = \frac{d \log D(t)}{d \log t}$$

Values of E(T) vs. log t are plotted in Figure 2. The vertical shift factors $V_1(T)$ and $V_2(T)$ for relaxation are taken to be $V_1 = h_2^{-1}$ and $V_2 = h_1^{-1}$ where h_1, h_2 are vertical shift factors for creep. Since ref. [1] provides only the product $h(T) = h_1(T)h_2(T)$, we take $V(T) = h(T)^{-1}$. The horizontal shift factor $a(T;\sigma)$ was approximated by the following expression: $a(T;\sigma) = a_T(T)a_\sigma(\sigma)$ where $a_\sigma(\sigma) = \exp\left[-\overline{\alpha}(\tau-\overline{\tau})\right]$ for $\tau > \overline{\tau}$ and $a_\sigma(\sigma) = 1$ for $\tau \le \overline{\tau}$. $a_T(T)$ was determined from a set of values $\theta_1 = 3.3$, $\log a_{T_1} = 6.0$; $\theta_2 = 2.69$, $\log a_{T_2} = 2.5$; $\theta_3 = 2.1$, $\log a_{T_3} = -8$ and $\theta_4 = 1.6$ $\log a_{T_4} = -10$ with $\theta_i = 1000/(273.1 + T_i \, °C)$. Intermediate values were obtained by linear interpolation. The parameter $\overline{\alpha} = \overline{\alpha}(T)$ was obtained by a spline function fit of $\overline{\alpha}(24.41) = 0.118$, $\overline{\alpha}(44.26) = 0.118$, $\overline{\alpha}(74.41) = 0.118$, $\overline{\alpha}(91.32) = 0.146$, $\overline{\alpha}(103.1) = 0.171$ and $\overline{\alpha}(120) = 0.239$. $\overline{\tau}(T)$ was obtained by a spline function fit of $\overline{\tau}(20) = 31.42$, $\overline{\tau}(26) = 31.3$, $\overline{\tau}(38) = 31.03$, $\overline{\tau}(50) = 30.8$, $\overline{\tau}(86.2) = 30$, $\overline{\tau}(108) = 22.16$, $\overline{\tau}(114) = 20$, and $\overline{\tau}(120) = 17.86$. The vertical shift V(T) corresponded to log V(40) = 0.025, log V(129.4) = -0.0031, and log V(200) = -0.21 with linear interpolation for intermediate values.

Finally, the coefficients of thermal expansion were taken to be $\alpha_L = 0.5 \times 10^{-6}/1°C$, $\alpha_T = 30 \times 10^{-6}/1°C$ for T < 125°C and $\alpha_L = 10^{-6}/1°C$, $\alpha_T = 75 \times 10^{-6}/1°C$, for 125°C < T < 300°C. To avoid computational difficulties at T = 125°C we assumed smooth transitions in values of α_L and α_T to occur over 124°C<T<126°C.

2.3 Optimal Cool-Down in the Linear Case

The optimization of cool-down can be stated as follows: Given an initial temperature T_I, where the composite is stress-free, a final temperature T_F ($T_F < T_I$), a finite time interval t_f, and a thermoviscoelastic stress-strain relation (e.g. equation (4)), find the "best" time-temperature history $T = T_\Omega(t)$, which minimizes the residual thermal stresses $\sigma(t_f)$. In all previous studies [6]-[9], which concerned linear behavior, namely a = a(T) in equation (5), it was found that $T_\Omega(t)$ undergoes sharp drops at t = o, from T_I to T_o and at t_f, from $T_\Omega(t_f)$ to T_F, and it varies monotonically and smoothly over the range o < t < t_f. Specifically for the linear case expressed in equation (4) (namely a = a(T) ≠ a(T;σ)) the initial drop is given by

$$T_o - T_I = \frac{V_1(T_o)a(T_o)}{a'(T_o)V_1(T_o) - V_1'(T_o)a(T_o)} \qquad (6)$$

which is a transcendental equation in the unknown T_o. Beyond the initial drop, the optimal path $T_\Omega(t)$ is governed by the integro-differential equation

$$\frac{d\,T_\Omega(t)}{d\,t} = -\frac{E''(t,t_f)}{E'(t,t_f)}\ \frac{a'\left[V_1'(T-T_I)+V_1\right]}{a\left\{a''\left[V_1'(T-T_I)+V_1\right] - a'\left[V_1'(T-T_I)+V_1\right]'\right\}} \qquad (7)$$

In equation (7), $E(t,t_f) = E\left(\int_t^{t_f} \frac{ds}{a(T(s))}\right)$ and primes denote derivatives with respect to the argument. Note that equation (7) must be solved "backwards" from $t = t_f$, towards $t = o$ since $E(t,t_f)$ cannot be evaluated without a-priori knowledge of the solution, with the exception of $E(t_f,t_f) = E(o)$ - which is known. The procedure is to guess $T_\Omega(t_f)$, solve equation (7) numerically and evaluate $T(o)$, then adjust the guess value of $T_\Omega(t_f)$ iteratively until we obtain $T(o) = T_o$ which matches the root of equation (6).

2.4 Non-Linear Optimal Cool-Down

(a) The Three Element Model as a Prototype Case.

Consider first the linear viscoelastic case with $E(t) = C_o + D_o \exp\ (-t/\lambda)$, $a(T) = \exp\left(-T/A + B\right)$ and $V_1 = V_2 = 1$. Straightforward manipulations yield [6]:

$$T_o = T_I - A, \qquad T_\Omega(t) = A\left[B - \ln\phi(t)\right] \qquad (8)$$

where $\phi(t) = t/\lambda + \exp C$, and $C = B+1 - T_I/A$. The residual thermal stresses during optimal cool-down are given by

$$(1-v)\,\sigma(t)/\alpha = A\left\{C_o\left[\ln\phi(t) + T_I\,/A - B\right] + D_o\right\} \qquad (9a)$$

or

$$\sigma(t) = -k\,T_\Omega(t) + k\,T_I + m\,A \qquad (9b)$$

where $k = \alpha E(\infty)/(1-v)$, $m = \alpha E(o)/(1-v)$. Introduce non-linearity by considering

$$a = a(T,\sigma) = \exp\left(-T/A + B - \beta\,\sigma\right) \qquad \beta > o \qquad (10)$$

5

The optimal cooling-path can be obtained by iteration, using the linear results (8) and (9) as initial guesses which are denoted by $A^{(o)}$, $B^{(o)}$, $T_\Omega^{(o)}$, $\sigma^{(o)}$. Namely, insert the linear result (9) into equation (10). After straightforward manipulations we obtain

$$a = (T,\sigma) = \exp\left(-T/A^{(1)}+B^{(1)}\right) \tag{11}$$

where $A^{(1)} = A^{(o)}/(1-\beta k A^{(o)})$, $B^{(1)} = B^{(o)}\left[1-\beta(kT_I+mA^{(o)})/B^{(o)}\right]$ with $A^{(o)} = A$, $B^{(o)} = B$. The primary significance of expression (11) is that it no longer contains the stress σ explicitly, hence we can write $a(T,\sigma) = a^{(1)}(T)$, and $a^{(1)}(T)$ is of the same form as $a(T)$. Consequently, $T_\Omega^{(1)}(t)$ and $\sigma^{(1)}(T)$ will retain the same forms as (8) and (9), but with $A^{(1)}$ and $B^{(1)}$ replacing A and B. At this stage, we substitute $\sigma^{(1)}(t)$ into equation (10) and obtain a revised $a(T,\sigma)$ which, as can be readily seen, is of the form $a^{(2)}(T) = \exp\left(-T^{(1)}/A^{(2)}+B^{(2)}\right)$ where $A^{(2)} = A^{(1)}$ and $B^{(2)} = B^{(o)}\left[1-\beta(kT_I + mA^{(1)})/B^{(o)}\right]$. An additional substitution yields $A^{(3)} = A^{(2)}$ and $B^{(3)} = B^{(2)}$ indicating convergence after three iterations. The optimal path for the three element model with viscoelastic non-linearity introduced through expression (10) is

$$T_\Omega(t) = \widehat{A}\left[\widehat{B} - \ln\widehat{\phi}(t)\right] \quad , T_\Omega(o^+) = T_I - \widehat{A} \tag{12}$$

where $\widehat{A} = A/(1-\beta k A)$, $\widehat{B} = B\left[1-\beta(kT_I+m\widehat{A})/B\right]$, and $\widehat{\phi}(t) = t/\lambda + \exp\left(1+\widehat{B}-T_I/\widehat{A}\right)$. The analytic solution (12) and the iteration scheme serve as a model and a verification check for the numerical scheme which follows.

(b) Optimal Cool-Down of [0/90]s APC-2 Composites.
The optimal temperature path was obtained numerically, by means of a special-purpose, custom-made computational sub-routine. The procedure was as follows:

Step 1: Consider linear viscoelastic behavior, with response functions $E(t),\alpha,a(T)$ and $V(T)$ determined from reduced experimental data and with prescribed T_I, T_F and t_f. Employ equation (6) to determine $T_o = T(o^+)$ and solve (7) numerically to obtain $T_\Omega(t)$, where $T_\Omega(t_f)$ is chosen iteratively until a solution of (7) yields $T_\Omega(o^+) = T_o$. Denote $T_\Omega(t)$ by $T_\Omega^{(o)}(t)$.

Step 2: Compute the residual thermal stress $\sigma(t)$, denoted by $\sigma^{(o)}(t)$, due to cool-down along $T_\Omega^{(o)}(t)$.

6

<u>Step 3</u>: Relate $\sigma^{(o)}(t)$ to $T_\Omega^{(o)}(t)$ to form $\sigma^{(o)} = F^{(o)}(T^{(o)})$.

<u>Step 4</u>: Consider the non-linear shift factor function $a(T;\sigma)$ which corre-sponds to the reduced data for APC-2. Along the linear optimal path this function is $a(T^{(o)}; F^{(o)}(T^{(o)}))$ which can be expressed as $a^{(1)}(T)$.

<u>Step 5</u>: Repeat steps 1, 2, 3, and 4 with $a(T)$ replaced by $a^{(1)}(T)$ to obtain $T_0^{(1)}$ with $T_\Omega^{(1)}(t)$, $\sigma^{(1)}(t)$, $\sigma^{(1)} = F^{(1)}(T^{(1)})$ and $a^{(2)}(T)$ respectively.

Continue until the attainment of a prescribed convergence, say

$$T_\Omega^{(n+1)}(t) = T_\Omega^{(n)}(t) + \epsilon(t) \quad , \quad |\epsilon(t)| < \text{ a given tolerance.}$$

Note the following observations:

1. The solution of equation (7) is, in general, unstable numerically. The optimal path undergoes rapid drops over short time-intervals, yielding vast variations in $a(T)$. Consequently, it is necessary to employ non-uniform time intervals when integrating equation (7).

2. The numerical correlation $\sigma = F(T)$ and its incorporation into $a(T;\sigma) \rightarrow a(T;F(T)) \rightarrow \hat{a}(T)$, as indicated in steps 3 and 4 above, require smoothing operations to provide reliable values for the derivatives of $\hat{a}(T)$ to be used in equation (7).

3. We have no mathematically grounded proof that the iterated optimization scheme must converge, and obviously no proof of uniqueness. For the APC-2 data at hand, convergence was attained after about 4-5 iterations. The validity of our results was verified at least partially by introducing several arbitrary, small disturbances in the optimal path and comparing $\sigma(t_f)$. In all cases the resulting values of $\sigma(t_f)$ exceeded the optimal value.

3. RESULTS AND CONCLUDING REMARKS

Results for optimal cool-down paths, $T_\Omega(t)$ vs. t, are shown in figures 3 and 4 for a cooling time $t_f = 100$ min with initial, stress-free, temperatures $T_I = 250°C$ and $300°C$, respectively, and $T_F = 30°C$. These figures contain results for the three thermorheological sub-cases: the "simple" case $(V = V_1 = V_2 = 1)$ and two "complex" cases $(V = V_1, V_2 = 1$ and $V = V_2, V_1 = 1)$, as well as three analogous non-linear cases with $a = a(T;\sigma)$. Figures 5 and 6 exhibit residual stress build-ups

during optimal cool-downs which correspond to the six sub-cases shown in Figures 3 and 4.

Figures 3-6 demonstrate the important role played by the various material functions which relate the thermoviscoelastic behavior of APC-2. Variations among those functions, as well as uncertainties concerning the stress free temperature lead to predictions of residual thermal stress which vary between 9.1 and 13.15 ksi, the ultimate transverse stress at room temperature being about 11.5 ksi. It is therefore desirable to extend the experimental characterization work of Xiao [1] to include a higher range of temperatures as well as transient temperature response to distinguish between the functions $V_1(T)$ and $V_2(T)$.

Acknowledgement

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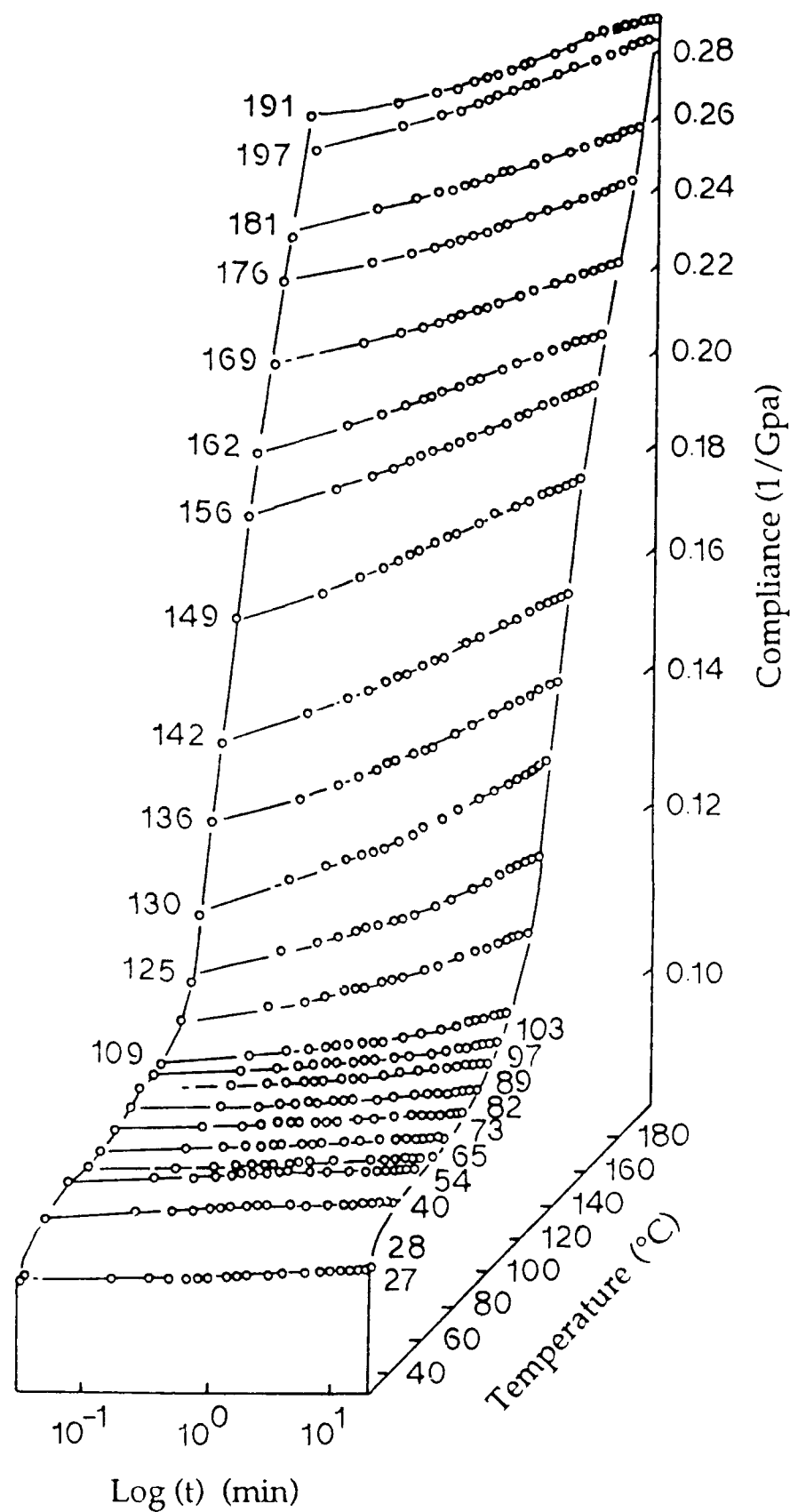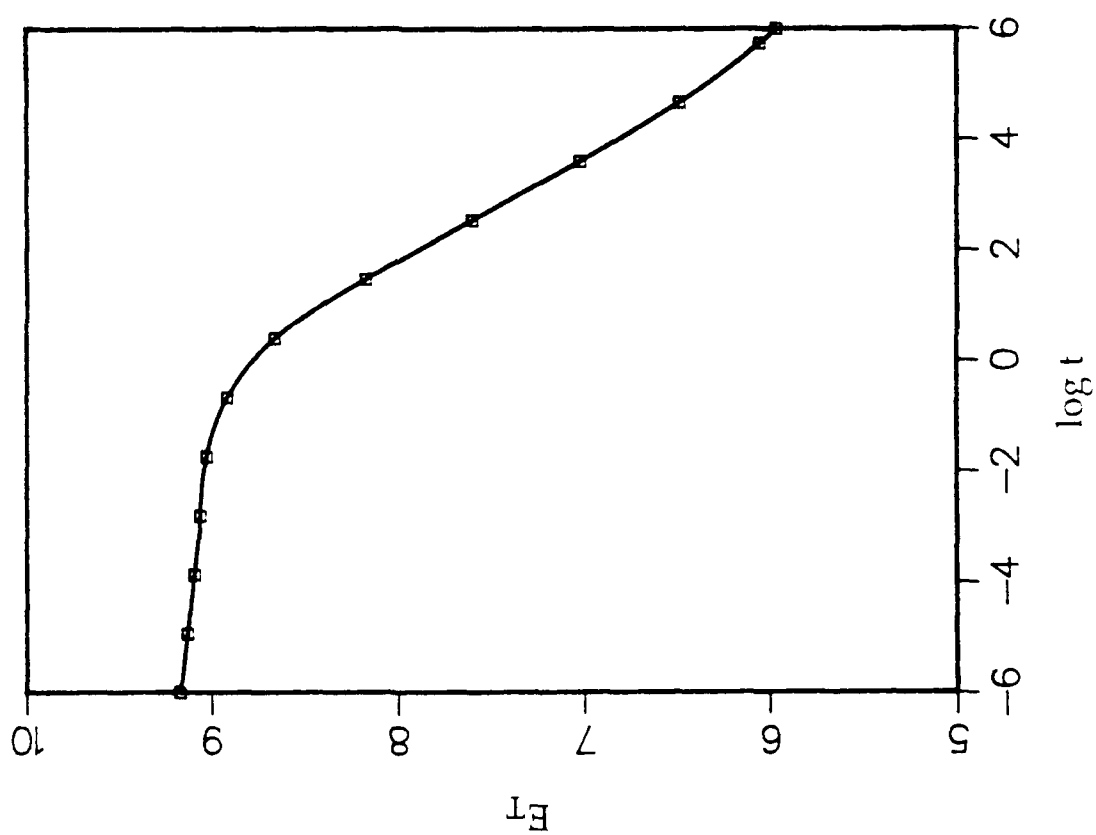
Figure 1.   Temperature dependence of transverse creep compliance of
APC-2 unidirectional laminate, at 15.22Mpa.   After Ref. [1].
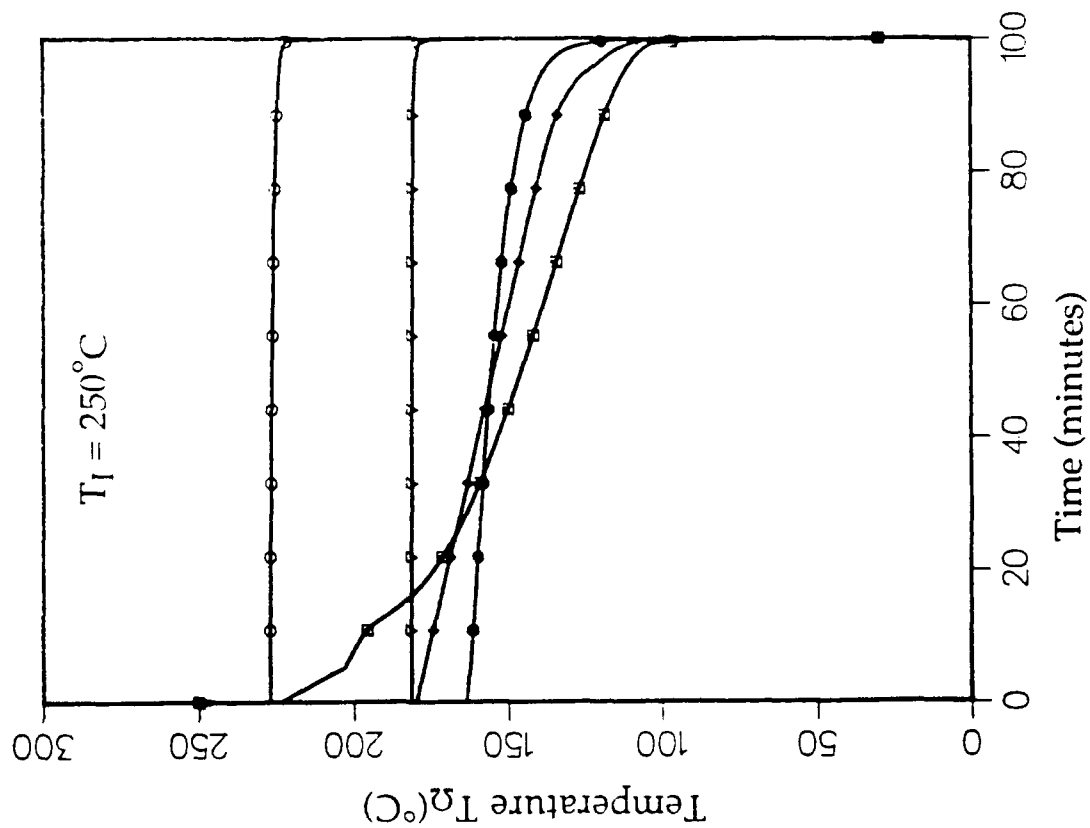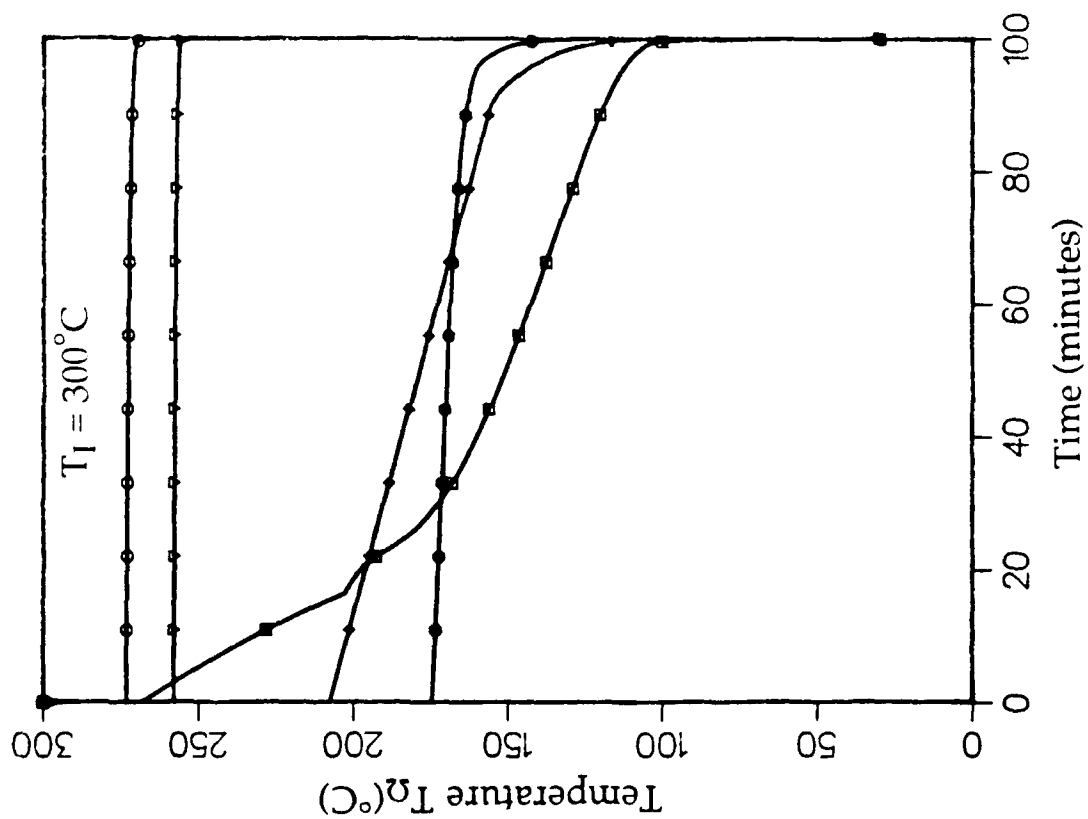
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Figure 2. The Relaxation Modulus E(t) vs. log t.

Optimal Temperature Paths $T_\Omega(t)$.

Residual Thermal Stresses Along The Optimal Temperature Paths

(a) With Horizontal Shift Only: Linear —□—□—, Non-Linear —+—+—,
(b) With Horizontal and Vertical Shifts (V = V_2): Linear —○—○—○—, Non-Linear —▽—▽—▽—
(c) With Horizontal and Vertical Shift (V = V_1): Linear —△—△—, Non-Linear —◇—◇—◇—.

Note that for linear behavior $T_\Omega(t)$ of cases (a) and (c) coincide.

Figure 3

Optimal Temperature Paths $T_\Omega(t)$.

Residual Thermal Stresses Along The Optimal Temperature Paths

(a) With Horizontal Shift Only: Linear —⊟—⊟—⊟—, Non-Linear —+—+—, (b) With Horizontal and Vertical Shifts (V = V_2): Linear —o—o—o—, Non-Linear —▽—▽—▽ (c) With Horizontal and Vertical Shift (V = V_1): Linear —◇—◇—◇—, Non-Linear —◇—◇—◇.

Note that for linear behavior $T_\Omega(t)$ of cases (a) and (c) coincide.

Fig. 4

Residual Thermal Stresses Along
The Optimal Temperature Paths

(a) With Horizontal Shift Only:
Linear ⊟⊟⊟ , Non-Linear ─+─
─+─,
(b) With Horizontal and Vertical
Shifts (V = V₂): Linear ─○─○─○─ ,
Non-Linear ▽ ▽ ▽
(c) With Horizontal and Vertical
Shift (V = V₁): Linear ─△△△
Non-Linear ◇ ◇ ◇.

$T_1 = 250°C$

stress σ (ksi)

Time (minutes)

Figure 5

14

Residual Thermal Stresses Along
The Optimal Temperature Paths

(a) With Horizontal Shift Only:
Linear —□–□–□—, Non-Linear —+
—+—;
(b) With Horizontal and Vertical
Shifts (V = V₂): Linear —o–o–o—,
Non-Linear —▽–▽–▽—
(c) With Horizontal and Vertical
Shift (V = V₁): Linear—△–△–△—,
Non-Linear –◇–◇–◇–.

Figure 6

15